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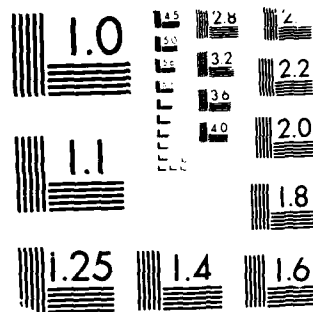
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Technical Report PTR-87-6

SDIO/IST ULTRASHORT WAVELENGTH LASER

**"Novel Experimental Schemes for Observing the
Mossbauer Effect in Long-Lived Nuclear Levels"**

By

Gilbert R. Hoy, Principal Investigator

Final Report
For the period ended March 31, 1987

Prepared for the
Naval Research Laboratory
4555 Overlook Ave., SW
Washington, D.C. 20375-5000
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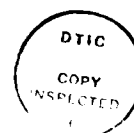
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By

Gilbert R. Hoy*

ABSTRACT

The development of gamma-ray lasers (GRASERS) will, in all probability, depend on the utilization of recoillessly emitted gamma rays from nuclear transitions, i.e. the Mossbauer effect (ME). Past research in Mossbauer spectroscopy which relates to GRASERS is briefly reviewed. The nuclear lifetimes, required from practical considerations, may have to be on the order of seconds, if storage and transfer processes prove infeasible. It is not clear that the ME has been observed in such long-lived states. Even if such long-lived states are not needed directly for GRASERS, successful observation of the ME in such a system will answer important questions of line broadening due to field inhomogeneities in single crystals. We propose some novel experimental schemes to observe the ME in ^{109}Ag whose relevant lifetime is about 40 seconds. The techniques proposed are: coincidence Mossbauer spectroscopy, conversion-electron Mossbauer spectroscopy, gravitational line sweeping, and gamma-ray self absorption. Preliminary results using the last technique are presented. Possible future, relevant ME research is discussed.

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1. INTRODUCTION

For approximately twenty years a small but dedicated group of researchers has been exploring the possibility of developing a gamma-ray laser (GRASER). A reasonably up-to-date analysis of the progress and problems in this area of research is found in the paper by Baldwin et al. (Ref. 1). In order to gain some perspective on the relationship of the Mössbauer effect (ME) to GRASER development, it is important to review the ME literature. The literature in this general field, we will call gamma-ray optics, starts in 1960. We have picked out particular results that we believe are quite significant, and will give a brief, heuristic description of each one.

1) Rayleigh and nuclear resonant scattering are coherent (Refs. 2-5). This result is, perhaps, surprising when one considers the processes involved. Rayleigh scattering occurs when the photon is elastically scattered from the Mössbauer atom as a whole. This so-called potential scattering is a "fast" process. On the other hand, nuclear resonant scattering is a "slow" process since, after the nucleus absorbs the photon, re-emission occurs on a time scale dictated by the nuclear lifetime.

2) One can obtain pure nuclear resonant diffraction by scattering from a sample for which the total Rayleigh scattering is very small (Refs. 6, 7). In this case one looks for a crystal in which the Rayleigh scattering from the Mössbauer atom is cancelled by destructive interference with Rayleigh scattering from other atoms in the unit cell.

3) Interference can occur between nuclear hyperfine components in nuclear resonant diffraction (Ref. 8). If one considers a single-crystal sample in which the resonant nuclei experience a finite hyperfine interaction, the resonant scattering cross-section will have a form containing a number of peaks corresponding to the number of allowed transitions, e.g.

usually six in the case of ^{57}Fe . If one does a resonant scattering experiment using such a sample, the resulting data will reflect this "split" pattern. Since the individual line shapes are essentially Lorentzian, some "lines" may overlap. In this region of the spectrum one must calculate the result by using a coherent superposition of the components.

4) In nuclear resonant Bragg scattering, processes having different final nuclear spin projections are incoherent (Ref. 9). This is simply the case where two scattering processes are distinguishable because the final states are different.

5) There can be anomalously deep penetration and high reflectivity using single crystals as a result of the "suppression of inelastic channel" (Ref. 10-16). The suppression of inelastic channels arises in single crystals as a consequence of the formation of eigenmodes of the radiation field. In certain cases the radiation field can be such that at the location of the Mossbauer nucleus, the amplitude for formation of the excited states is zero. If the nucleus is not excited, it cannot de-excite through the inelastic channel of internal conversion.

In addition to the work noted above, there has been a considerable related theoretical effort made by G. T. Trammell, J. P. Hannon and collaborators (Refs. 17-21). Some recent experiments have involved the use of synchrotron radiation (Refs. 22-26). In these experiments Mossbauer diffraction was observed using a synchrotron as the photon source. One important result (Refs. 22, 24-26) was that the decay of the resonantly excited nuclei in the single crystal along Bragg directions was more rapid than expected from the value of the nuclear lifetime. The same effect was also seen in ordinary nuclear resonant scattering experiments (Ref. 15) by observing that in the scattered spectrum, in the Bragg direction, the lines

were about fifteen times the natural linewidth that is predicted by the time-energy Heisenberg relation. Subsequent experimental results have dealt with further amplification of the consequences of the effects mentioned above concerning coherent effects in nuclear resonant diffraction (Refs. 27-35).

Even though gamma-ray optics has perhaps not yet emerged as a field in its own right, the brief review given above shows that some very significant results have already been obtained. In what follows we focused more on ME studies directly related to possible GRASER development. One of the important issues is how narrow can the linewidth of a recoillessly emitted gamma ray actually be. A related question concerns the degree of field homogeneity at the nucleus that can exist in single crystals. Exploration of the ME in long-lived states will answer some of these fundamental questions.

The first excited states of ^{107}Ag and ^{109}Ag each have lifetimes of about 40 seconds and corresponding linewidths of approximately 1×10^{-17} eV; see Fig. 1. Such transitions provide an obviously severe test for the observance of the ME and hence may shed some light on the linewidth and field inhomogeneity questions. Three short papers (Refs. 36-38) exist which purport to have observed the ME in these transitions. It is fair to say that the scientific community at large has been skeptical of these assertions. We proposed some possible experimental techniques which, if successful, would confirm the observation of the ME in ^{109}Ag . This isotope was selected because of the favorable lifetime (460 days) of the parent ^{109}Cd . The methods proposed are: coincidence Mössbauer spectroscopy (Refs. 39-46), conversion-electron Mössbauer spectroscopy (Ref. 47), gravitational line sweeping based on the gravitational red shift (Ref. 48), and temperature

dependence of resonant self absorption. Preliminary results are presented below using the temperature dependence of self-absorption method.

Finally, we discuss some future ME-type measurements related to GRASERS and gamma-ray optics in general.

2. PROPOSED EXPERIMENTAL TECHNIQUES

The coincidence Mössbauer spectroscopic technique is very similar to that of perturbed angular correlation and nuclear lifetime measurement methods (Refs. 49, 50). The $t = 0$ pulse, in the present case, could be due to an x ray produced following the electron capture in cadmium leading to the formation of the silver first excited nuclear level. The gamma ray resulting from the nuclear decay to the ground state is then counted, after passing through the rest of the silver sample, in delayed coincidence with the x ray.

The theory for the transmission of the electromagnetic wave through a Mössbauer resonant absorber as a function of time for this coincidence Mössbauer case has been worked out by Hamermesh (Ref. 41). The point here is that the measured decay curve can differ markedly from the typical lifetime exponential shape. (In fact, using ^{57}Fe the experiment is easy (Ref. 51) and the results are dramatic, see Fig. 2.) The deviation from the exponential shape depends on: Γ the linewidth, the frequency difference between the emission and absorption lines (assumed zero for our purposes), and the "Mössbauer" thickness of the absorber.

A second promising method is to look at the conversion electrons emitted after the resonantly absorbing nuclei are excited (if they are). The internal conversion coefficient, α , is approximately 20 for silver. Thus conversion electron Mössbauer spectroscopy (CEMS) is quite feasible. As

long as the conversion electrons from the source portion of the sample are sufficiently attenuated so that no primary source conversion electrons can be detected, the signal-to-noise ratio of this experiment is several orders of magnitude greater than in ordinary Mossbauer spectroscopy.

If it becomes clear that the ME does occur in silver, the gravitational line sweeping method may be used to actually measure the lineshape. In this case one could do CEMS using a sample and an experimental set-up schematically represented in Fig. 3. The whole configuration can be rotated about an axis perpendicular to the gravitational field. A simple calculation using ^{109}Ag shows that if the gamma-ray photon falls $\sim 1 \times 10^{-4}$ cm in the gravitational field, its energy is raised by about one linewidth. If the experimental parameters are arranged so that one primarily detects events corresponding to γ rays that pass from the source side "straight" toward the detector side, rotation of the apparatus should bring more absorber nuclei into resonance. The number of conversion electrons detected should thus be a function of the angle ϕ . One could easily use a multichannel analyzer in a mode where each channel corresponds to a particular angle. Rotating the crystal back and forth would "sweep" through the resonance.

3. PRELIMINARY RESULTS ON ^{109}Ag USING GAMMA-RAY SELF ABSORPTION

For the ^{109}Ag experiments, one millicurie of carrier free ^{109}Cd was purchased from New England Nuclear along with a single crystal of silver purchased from Monocrystals Co. This single crystal is in the form of a disk having a nominal half inch diameter and a thickness of 0.4 mm. It was grown in the (111) orientation. Silver is 48.7% ^{109}Ag and 51.3% ^{107}Ag .

A special cell was designed so that the ^{109}Cd could be electroplated onto a face of the silver single-crystal specimen. After the ^{109}Cd had been deposited on the single crystal, the sample was placed in a tube furnace for

annealing. The sample was slowly warmed in an argon-hydrogen atmosphere, then annealed for one hour at 400°C and for six hours at 350°C. Because the Cd has a rather high vapor pressure at 400°C, care had to be taken to avoid having ^{109}Cd vapor deposited on the backside, i.e. the non-plated side of the single-crystal specimen.

The spectrum of the ^{109}Cd was determined by using an intrinsic germanium solid-state detector. The spectrum consists of: silver K_α and K_β x rays, the 88-keV gamma ray, the associated escape peaks, scattered radiation, and Compton background. The spectra of the single-crystal specimen were taken from both the "front" and "back" side of the sample before and after each stage of the annealing process. The mass absorption coefficients of silver metal are 14, 10.5, and $2.02\text{ cm}^2/\text{g}$, respectively, for photon energies of 22, 25, and 88 keV. The density of silver at room temperature is 10.5 g/cm^3 . Using these numbers and our measured counting rates from the front and back sides of the sample, it was determined that the ^{109}Cd diffused into the silver to an effective average depth of approximately $94 \pm 2\text{ }\mu\text{m}$ ($\sim 3.7 \times 10^{-3}$ inches). Figure 4 gives a schematic representation of the sample.

Before attempting to do the more elaborate experiments mentioned above, it was decided to try first to observe the ME itself in a fairly straightforward manner. If one observes the pulse-height spectrum of the sample from the "back side" using the Ge solid-state detector as a function of temperature of the sample, it should be possible to discern any ME. The idea is as follows. Since the recoilless fraction (f) in silver using the 88-keV, γ -ray transition in ^{109}Ag is $\sim 3.5\%$ at liquid helium temperature and only 0.3% at liquid nitrogen temperature (Fig. 5 shows the temperature dependence of f), one would expect that, due to Mössbauer self-absorption

the number of gamma rays would decrease and the number of x rays, following internal conversion, would increase as the temperature decreases. Thus, the ratio of the number of x rays to γ rays reaching the detector after passing through the remaining portion of the sample should be higher at liquid helium temperature than at liquid nitrogen temperature. Using the ratio circumvents most of the problems associated with detector stability, electronic drifts, and source decay.

Figure 6 shows the pulse-height spectrum of our sample containing ^{109}Cd diffused into the silver single crystal. This spectrum was taken from the back side with the sample at 75 K. We define three regions of interest: one containing the K_α x-ray contribution, the second for the K_β contribution, and the third for the γ -ray contribution. We define $R_{\alpha\gamma}$ to be the ratio of the number of counts in the K_α x-ray portion to that of the γ -ray portion, and $R_{\beta\gamma}$ to be a similar ratio for K_β to the γ ray.

For a given experimental set-up it is possible to calculate the values of $R_{\alpha\gamma}$ as a function of temperature and "effective Mössbauer" thickness x_N . Such a calculation thus parameterizes the result in terms of the effective Mössbauer thickness. The result, neglecting specific experimental details for clarity, is

$$R_{\alpha\gamma} = \frac{I_{0\alpha} e^{-\mu_\alpha x}}{I_{0\gamma} [(1-f)e^{-\mu_\gamma x} + f e^{-(\mu_\gamma x + \mu_N x_N)}]} \quad (1)$$

where $I_{0\alpha}$ and $I_{0\gamma}$ are constants, μ_α is the absorption coefficient of silver for the K_α x rays (~ 22 keV), f is the recoilless fraction, μ_γ is the absorption coefficient of silver for the γ ray (~ 88 keV), μ_N is the nuclear resonant absorption coefficient for the γ ray, x is the thickness of silver in

the path, and x_N is the effective nuclear resonant thickness.

It should be noted that by taking the ratio, as expressed in Eq. (1), the decay of the source and solid angle considerations have cancelled out. Notice also that f , μ_Y , μ_a and μ_N are temperature dependent. The temperature dependence of f has already been shown in Fig. 5. The temperature dependences of μ_Y , μ_a , and μ_N occur because the density of silver enters in the absorption coefficient. All these effects can be included using the literature (Refs. 52, 53) data. The quantity $\mu_N = n_0 P \sigma_N$ where: n_0 is the number of silver atoms/cm³, P is the fraction of ¹⁰⁹Ag atoms in the sample, and σ_N is the Mössbauer resonant cross section on resonance. We took $\sigma_N \approx 4.57 \times 10^{-20} \times f \text{ cm}^2$.

Our initial experiments were done in the vertical geometry, i.e. the K_α x rays and the γ rays were "falling" through our sample into the Ge detector positioned below. The preliminary results are shown in Figs. 7a and 7b. The bottom solid curve in each figure is the expected result for R_{ar} according to Eq. (1) without the Mössbauer effect occurring. (The calculation was actually modified slightly because the "beam" path included aluminum in the sample holder and a beryllium window in the dewar.) The top solid curve through the data points in each figure is characterized by an "effective nuclear thickness." It should be noted that the theory and experiment were normalized at 75 K. The effective nuclear thickness that characterizes the rough fit to our data can be interpreted in the terms of an effective line broadening. Thus the "back side" result would suggest an effective Mössbauer resonant linewidth of about 24 times the natural linewidth while the "front side" gives a result of 16 times the natural linewidth. It is encouraging, but also somewhat surprising, that these

values seem to agree with each other. The data in Fig. 7a are not compellingly convincing while those in Fig. 7b perhaps only appear so, because we obtained less data. Since these data were taken, several months have been spent working in the horizontal geometry. We have been unable to obtain meaningful results, as yet, due to insufficient reproducibility in our experimental data.

4. FUTURE RELEVANT MÖSSBAUER EFFECT RESEARCH AND CONCLUSIONS

G. T. Trammell, J. P. Hannon, and collaborators have predicted the possibility of multibeam Borrmann modes arising from recoillessly emitted γ rays from nuclei deep inside single crystals. They have also predicted that such effects will have substantial practical advantage for GRASER development (Refs. 19-21, 54-56). We are considering various possible experimental techniques for observing these multibeam Borrmann modes. These include the use of microchannel plate detectors incorporating various commercial read-out mechanisms and schemes for looking at the single-crystal source radiation along certain directions using Ge solid-state detectors and interposing rotating, collimating, absorption screens containing prescribed patterns of holes. Figure 8 shows a possible schematic experimental arrangement for observing multibeam Borrmann modes.

A great deal of research has been done by a relatively small group of workers on nuclear resonant diffraction as noted above. This has involved the study of single crystals using the ME in scattering or transmission geometries. It appears that the so-called "suppression of inelastic channels" has been established. In addition, nuclear, coherent, macroscopic radiation states have apparently been prepared by illuminating single crystal samples with nuclear resonant radiation. Unfortunately no experiments

have been done on the study of single-crystal sources themselves. Research on this subject is vitally needed.

The status of the ME in ^{109}Ag is still cloudy although we have obtained some encouraging preliminary results. Our results roughly agree with the work of Wildner and Gonser (Ref. 38) in that we deduce an effective linewidth of approximately 20 times the natural linewidth. Further research is in progress.

Finally, we would like to close this report by listing several questions, sometimes provocative ones, that could benefit the GRASER project and possibly be answered by further Mössbauer studies. How perfect do the crystals really have to be? Can one produce narrow single-line sources in single crystals? Can radioactive nuclei be put in perfect single crystals having a high recoilless fraction and still give a narrow-line spectrum? What is the narrowest line possible? What way is best for producing these sources: ion implantation, chemical deposition, etc.? Are there rapid ways of converting non-single crystals into single crystals e.g. by laser annealing? Can one observe the multibeam Borrmann modes and then measure the resonant nuclear coupling coefficients? Do nuclear, coherent, macroscopic radiating states exist in single crystal sources themselves? Interestingly, this is not a new idea. Theoretical papers do exist on this subject (Refs. 57-62). Is it impossible to produce radioactive sources in the form of single crystals which will radiate in certain directions with a lifetime much shorter than the ordinary lifetime of the individual radioactive nuclei? The amount of Mössbauer effect research that could be done in connection with the GRASER project seems to be enormous. More importantly, it appears such research may produce important, exciting, new physics.

5. ACKNOWLEDGMENTS

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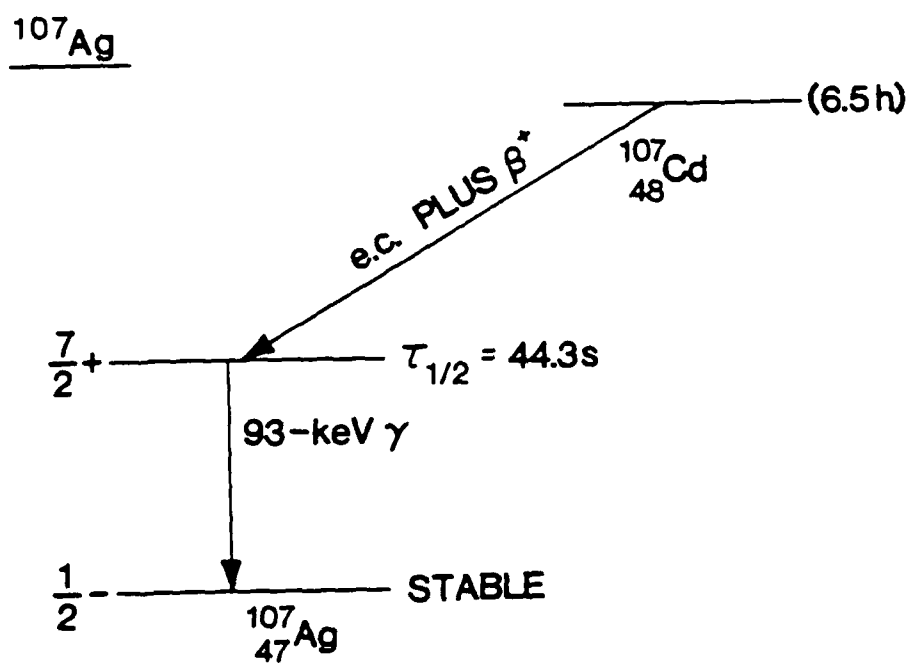
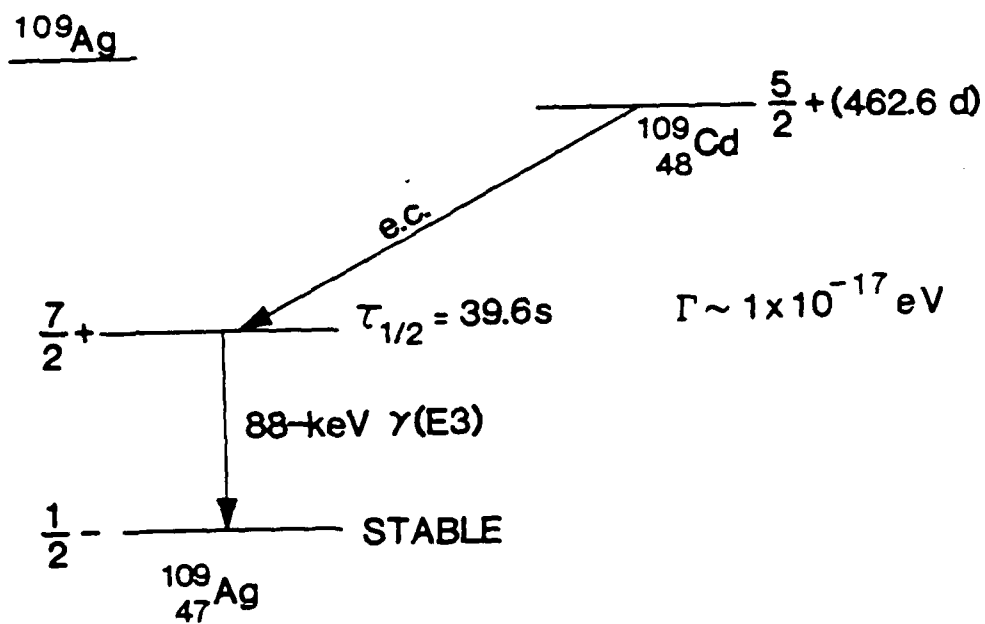


Fig. 1. The decay schemes for ^{109}Cd and ^{107}Cd .

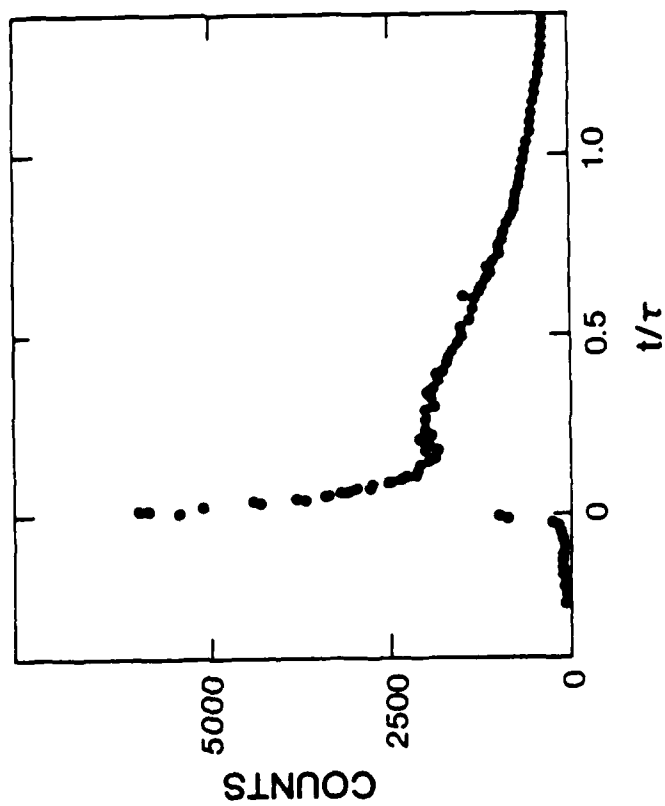


Fig. 2. A typical lifetime curve as measured through a ^{57}Fe nuclear resonant filter, which is on resonance, using a ^{57}Co source. Notice that the shape of the curve is much different from the usual exponential form.

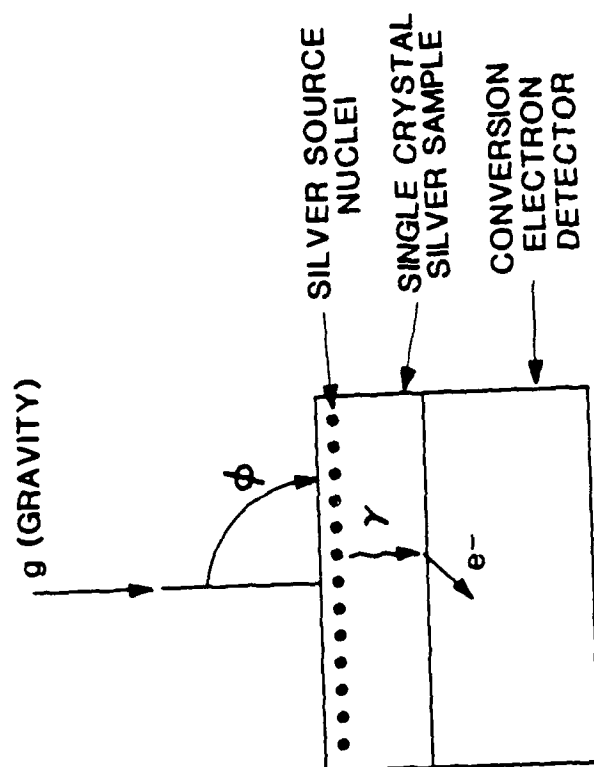


Fig. 3. Schematic representation of a possible experimental configuration for using the gravitational sweeping technique to measure the ^{109}Ag nuclear resonant line shape.

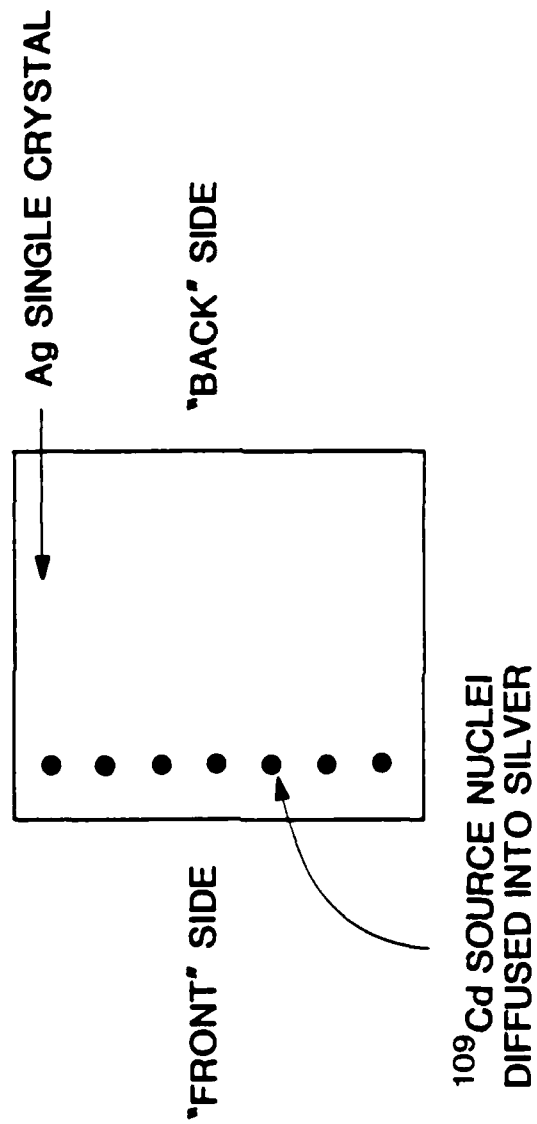


Fig. 4. Schematic representation of the ^{109}Cd in a silver single-crystal sample.

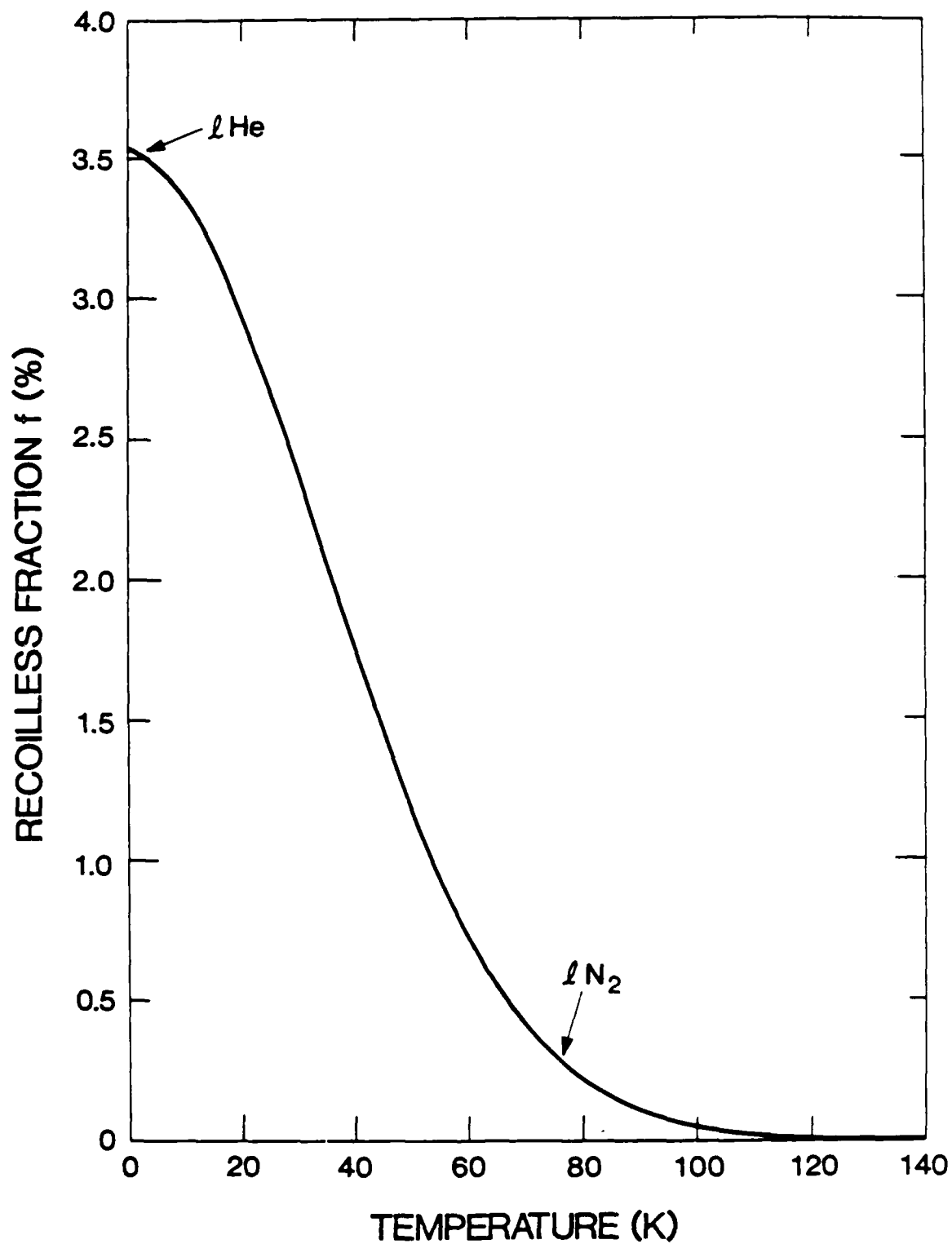


Fig. 5. The temperature dependence of the recoilless fraction (f) of the 88-keV transition in silver having a Debye temperature, $\theta_D = 226$ K.

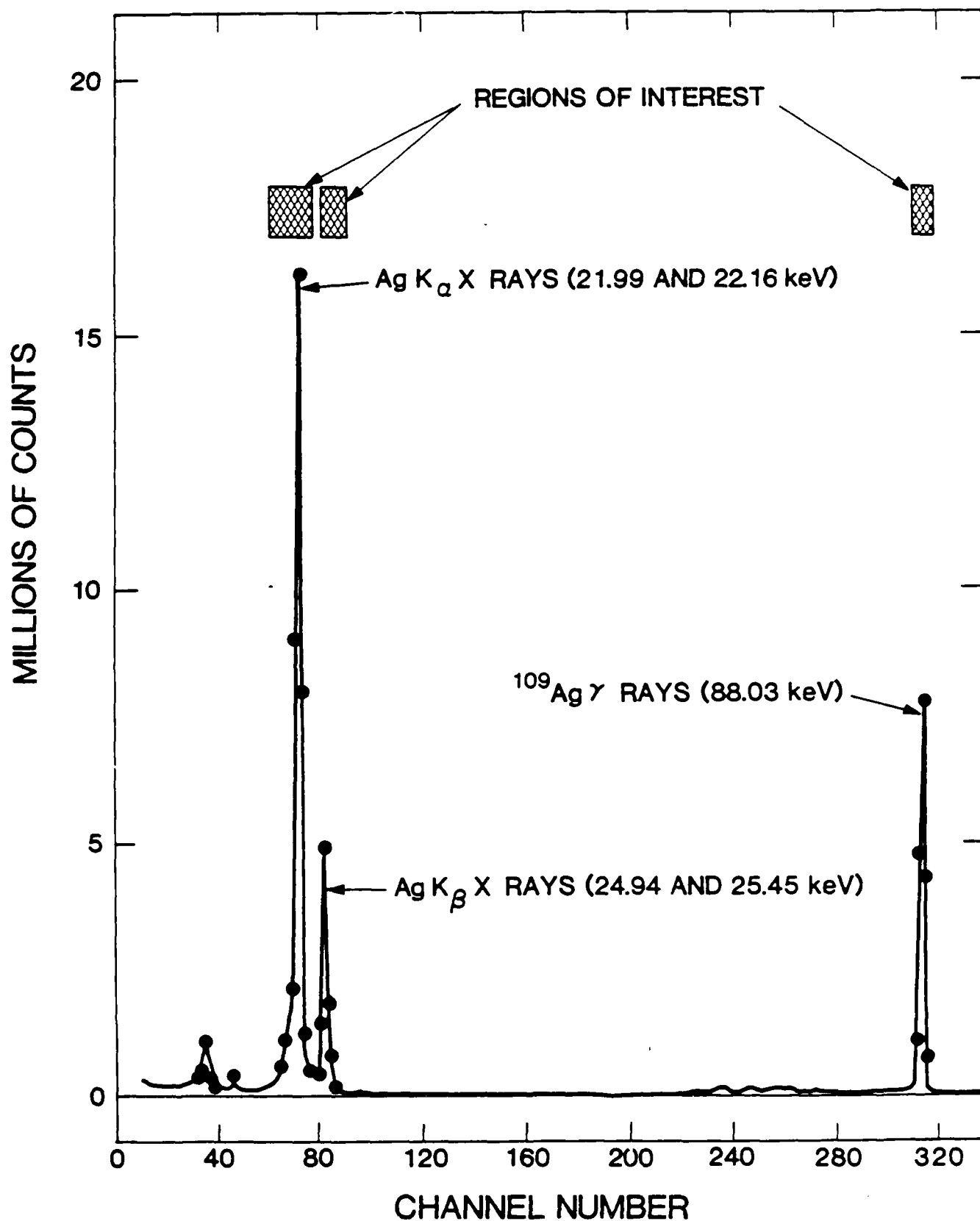


Fig. 6. Ge detector pulse-height spectrum of ^{109}Cd in the silver single-crystal sample. The spectrum was taken from the "back" side of the 0.4-mm thick sample at 75 K.

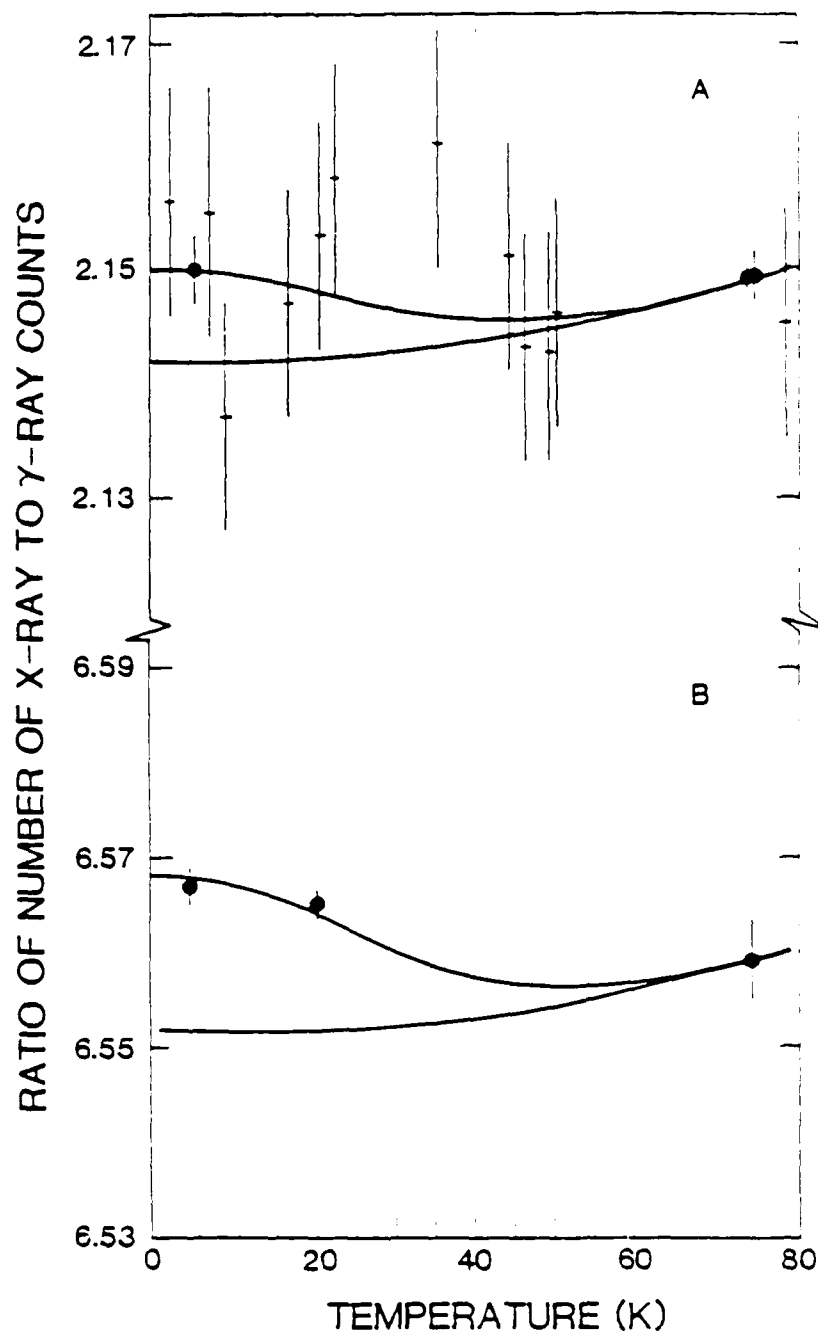


Fig. 7. a) Experimental and calculated results for the ratio R_{xy} measured in a vertical geometry from the "back" side of the sample as a function of temperature. The bottom solid curve gives the calculated results assuming no Mossbauer effect. The upper solid curve is the expected result assuming an effective nuclear thickness, $x_N = 2.4 \times 10^{-3}$ cm. Notice that the experimental data at temperatures of 4, 74, and 75 K have significantly better statistics than at the other temperatures. The theory and experiment were normalized at 75 K. b) Experimental and calculated results for the ratio R_{xy} measured from the "front" side of the sample as a function of temperature. The bottom solid curve gives the calculated results assuming no Mossbauer effect. The upper solid curve is the expected result assuming an effective nuclear thickness, $x_N = 1.6 \times 10^{-3}$ cm. The theory and experiment were normalized at 75 K.

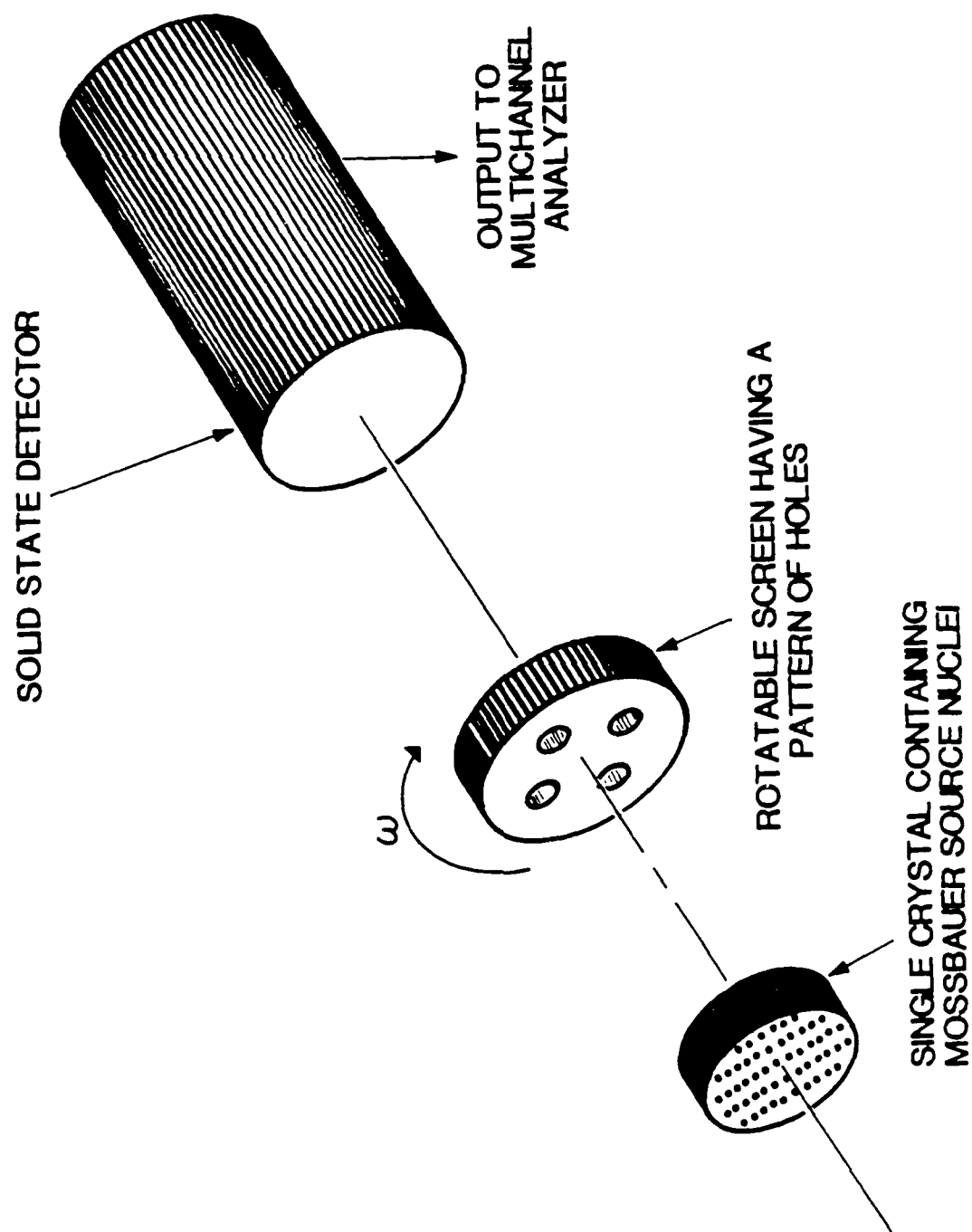


Fig. 8. Schematic representation of a possible experimental configuration to observe multibeam Braggman modes.

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